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Distribution and geochemical processes of boron in the multimedia of Lake Qinghai, China

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ABSTRACT

Lake Qinghai on the Qinghai-Tibet plateau is the largest lake in China. This study investigated the concentration and geochemical processes of boron (B) in lake water, lake sediment and river samples collected from Lake Qinghai and the Buha River. In addition, lake sediment pore water samples were analyzed. The concentrations of B and major ions, including K^+ , Na^+ , Ca^{2+} , Mg^{2+} , Cl^- and SO_4^{2-} , were analyzed in all the water samples. The average concentration of B was 0.07 mg L^{-1} , 6.37 mg L^{-1} , 12.79 mg L^{-1} and 59.42 mg kg^{-1} for river water, lake water, pore water, and sediment, respectively. There were significant ($p < 0.05$) and positive ($r = 0.70$) relationships between the B concentrations in bottom water of the lake and in lake sediment, indicating that B diffusion from the sediment plays an important role in the concentration of B in bottom lake water. The differences in B concentrations and B/major ion molar ratios of the river water, lake water, and pore water indicated the following geochemical processes: 1) B is enriched in the lake water through evaporative concentration; 2) B is removed from the lake water through mineral precipitation as well as sorption onto colloids; 3) Solid-phase B in sediments was released through dissolution driven by organic matter mineralization. B/Cl and Na/Cl molar ratios alone are not enough to identify the sources of B in the water of inland closed-basin saline lakes because of these processes.

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Introduction

Boron (B) is a naturally occurring element that is found in nature in the form of borates in the ocean, sedimentary rocks, coal, shale, and some soils. Natural sources of borates released into the environment include the oceans, geothermal steam, and natural weathering of clay-rich sedimentary rocks (Wedepohl and Correns, 1969). Boron is also released from anthropogenic sources to a lesser extent. Boron is an essential micronutrient for plants and animals. When the concentration of B $< 1 \text{ mg L}^{-1}$, the risk of adverse effects from boron on the aquatic ecosystem is low, but when the concentration of B is too high, aquatic organisms in such habitats may be affected (WHO, 1998).

Over the past several decades, significant research efforts have been made to study the distribution of B in surface water. The B concentrations for seawater and fresh water have been summarized. Reimann and Caritat (2012) reported that the worldwide median concentration of B in ocean water is 4.5 mg L^{-1} . Gaillardet et al. (2003) summarized the B concentrations in approximately 60 rivers around the world and found that the mean content of B in river water is 0.01 mg L^{-1} , ranging

from 0.002 to 0.15 mg L^{-1} . An extensive survey of trace metals in surface waters of the U.S. was conducted between 1962 and 1967. B was detected in 98% of the 1577 samples collected over this 5-year period, and the detectable B levels ranged from 0.001 to 5 mg L^{-1} , with a mean value of 0.1 mg L^{-1} (Butterwick et al., 1989). Wyness et al. (2003) presented the main characteristics of surface water B data for each of the European Union countries. The results indicated that the B concentrations vary considerably throughout Europe (from <0.001 to 7.49 mg L^{-1}), with the highest values occurring in Belgium, Germany, Greece and Portugal and the lowest concentrations occurring in Finland, Austria and the UK.

In contrast, the surface waters in Asia have received relatively little attention, except for a number of key rivers. Gaillardet et al. (2003) summarized the mean B concentrations in the Lena, Yangtze, Yellow, Ganges and Mekong rivers and found the concentrations of 4.7 , 12.5 , 150.0 , 17.8 and $15.0 \text{ } \mu\text{g L}^{-1}$, respectively. In China, the studies of B have focused on B isotope compositions of brines from different salt lakes in Inner Mongolia, Qinghai, Xinjiang and Tibet provinces in western China. Those salt lakes have unusually high concentrations of boron and the extraction of B from those salt lake brines are important economic resources for those areas (Fan et al., 2015; Tan et al., 2012; Wei et al., 2014). The results suggested that, due to the influence of evaporation and the surrounding water sources, the concentrations of B and its

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isotopic composition vary in different regions and even in different salt lakes in the Qaidam Basin of Qinghai Province (Qi et al., 1993; Xiao et al., 1992, 1999, 2000; Vengosh et al., 1995; Liu et al., 2000).

Qinghai Lake, the largest lake in China is located in a critical transitional zone between the humid region controlled by the East Asian monsoon and the dry inland region affected by westerly winds (Li et al., 2012). Due to its unique geological/geographic settings, Lake Qinghai catchment has become an outstanding, world-class site for scientific research and is attracting increasing attention (Xu et al., 2006; Colman et al., 2007; Jin et al., 2009, 2010a, 2010b). Xu et al. (2010) studied the major ion concentrations in the water of Lake Qinghai, showing that the chemical composition of the lake water can be mainly ascribed to evaporation and crystallization. Sun et al. (1993) reported on the B isotopic composition in the water of Lake Qinghai, and demonstrated that B is of continental genesis and is not related to seawater. The mean B concentration has been estimated to be 10.33 mg L⁻¹ in shallow water of Lake Qinghai, 10.29 mg L⁻¹ in semi-deep water of Lake Qinghai, and 0.74 mg L⁻¹ in the water of Buha River (Wang et al., 2017a, 2017b). Although a detailed understanding of the B concentrations in lake water, sediment and river water is crucial to better define geochemical processes of B within Lake Qinghai catchment, to date, there have been no comprehensive studies about B in Lake Qinghai catchment.

In this study, 69 lake water samples, 22 lake sediment samples, 21 pore water samples and 12 river water samples were collected systematically within Lake Qinghai catchment to determine the concentration of B in these media. The main objectives of this study were to determine the concentrations of B in lake water, sediment, and pore water in Lake Qinghai and influent river water, to investigate the vertical distribution of B concentrations in lake water, and to explore the relationship between the B concentration and other ions in lake water and possible interactions with the lake's sediments. Knowledge of the B geochemistry and geochemical evolution of water and sediment in these areas might be useful to better understand the hydrochemical systems and promote the sustainable development of water resources.

Materials and methods

Study site

Lake Qinghai (36°32'–37°14'N, 99°37'–100°45'E) lies in an intermountain basin. It is located in the northeastern Tibetan Plateau at an altitude of 3194 m above sea level (m.a.s.l) (Xiao et al., 2012). It is the largest inland lake (with a water area of 4260 km² and a catchment area of 29,660 km²) and the largest closed-basin saline lake (average salinity 14 practical salinity units) in China (Wang et al., 2017a, 2017b). The lake is relatively deep compared with other large lakes in China, with average and maximum water depths of approximately 21 and 29 m, respectively (Sha et al., 2017). Lake Qinghai catchment is characteristic of the Qinghai-Tibet Plateau continental climate, with a mean annual precipitation of 336.6 mm (1951–2005) and an average amount of evaporation of 925 mm (1959–2000) (Colman et al., 2007). Air temperature around the Lake Qinghai region varies between 10.4 and 15.2 °C in July, and between -10.4 and -14.7 °C in January, with a mean annual value of about -0.7 °C (Xu et al., 2006). The lake is temperature-stratified in summer (hypolimnion of 6 °C, epilimnion 12–15 °C) but this breaks down as a result of autumn overturn (Henderson and Holmes, 2009). 90% of the precipitation occurs between June and September, resulting in nearly 85% of riverine discharge during this period (Ding and Liu, 1993). There are approximately 40 rivers feeding the lake, but most are ephemeral. Buha River is the largest one followed by the Shaliu River and the Haergai River. These three rivers provide 42.6%, 15.3% and 15.0% of the total runoff quantity into the lake, respectively (Li et al., 2007; Li et al., 2009).

The sediment of pelagic regions of the lake usually consisted of 30% clay (illite, kaolinite, and chlorite), 28% quartz, 15% plagioclase, 13%

calcite, 10% aragonite, 2% K feldspar, and 2% dolomite; while the sediment of littoral areas generally consisted of 41% quartz, 21% plagioclase, 17% clay (illite, kaolinite, and chlorite), 12% calcite, 5% K feldspar, 3% dolomite, and 1% iron pyrite (Wang et al., 2017a, 2017b).

Lake and river water collection and analysis

Lake water samples were collected from 23 stations (Fig. 1), labeled L01–L23, in September 2016. Surface lake water was sampled approximately 2 m below the water surface for all the 23 sampling sites. For eight sample stations (L03, L04, L05, L08, L13, L14, L17 and L19), lake water samples from different depths (from 2 m to bottom in 4-m intervals) were collected with a 2-L polyethylene water sampler. River water samples were collected at 12 sites along Buha River.

The water samples were filtered immediately through a 0.45- μ m Teflon filter, acidified to pH 1.5–2.0 with HNO₃ and stored in pre-cleaned high-density polyethylene plastic bottles for B and water ion analysis. pH was measured at the field by pH meter (Horiba, Japan), which was calibrated by Merck pH buffer solutions (pH = 4.0, 7.0, and 10.0). The total alkalinity was determined using the acid-base titration method. The titrations were conducted in a room where temperature varied from 22 to 24 °C. 50 mL water sample was pipetted into a 250 mL Erlenmeyer flask. To this, 2–3 drops of methyl orange indicator solution (0.1%) was added. The sample was stirred with a magnetic stirrer (80 rpm). The endpoint for total alkalinity determination was signaled with methyl orange indicator; 0.025 M standard HCl titrant was used to titrate the solution and at the end point of the total alkalinity determination of water the indicator changes from yellow to faint orange (pH = 4.4).

The Cl⁻ and SO₄²⁻ in the water samples were measured using an ion chromatography system (ICS) (ICS - 1100, America). Concentrations of K, Na, Ca, Mg and B in the water were determined by inductively coupled plasma-mass spectrometry (ICP - MS) (X Series II, Thermo Electron).

Sediment collection and analysis

Lake surface sediment (approximately 0 to 20 cm) samples were collected from all 23 sites (L01–L23), except for L22, using cable operated sediment samplers-Van Veen grabs (Eijkelpkamp), and they were transferred to acid-washed, dark-colored polyethylene bags and transported to the laboratory, where they were freeze-dried, slightly crushed, passed through a 2 - mm sieve and stored in glass bottles until analysis. The sediment samples were digested with HNO₃ - HF - HClO₄ (Lin et al., 2007). The K, Na, Ca, Mg, S and B in the extracts were measured with inductively coupled plasma atomic emission spectrometry (ICP - AES) (IRIS Intrepid II, Thermo Electron).

Pore water collection and analysis

Sediment pore water was extracted from each sediment site (except for L16 and L23) by centrifugation in polyethylene tubes for 15 min at 12000 rpm at 4 °C (CL17R, America). The supernatant pore water was re-centrifuged again for 15 min to remove excess particulates, after which it was filtered through a 0.45 - μ m Teflon filter and stored in a polyethylene container in the dark at -20 °C until required for analysis. The analytical methods for the pore water samples were identical to the lake and river water methods.

Quality control

The accuracy of the method used to analyze the B in the sediments was determined by analyzing the B concentration of certified reference materials GSS21, which were provided by the Institute of Geophysical and Geochemical Exploration, Chinese Academy of Geological Sciences. The recovery of B for GSS21 ranged from 95.88% to 119.16% with an

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